



On the validity of the Geiger–Nuttall alpha-decay law and its microscopic basis

C. Qi^{a,*}, A.N. Andreyev^{b,c}, M. Huyse^d, R.J. Liotta^a, P. Van Duppen^d, R. Wyss^a

^a KTH, Alba Nova University Center, SE-10691 Stockholm, Sweden

^b Department of Physics, University of York, YO10 5DD, United Kingdom

^c Advanced Science Research Center, Japan Atomic Energy Agency (JAEA), Tokai-mura, 319-1195, Japan

^d KU Leuven, Instituut voor Kern-en Stralingsfysica, 3001 Leuven, Belgium

ARTICLE INFO

Article history:

Received 24 April 2014

Received in revised form 17 May 2014

Accepted 21 May 2014

Available online 27 May 2014

Editor: W. Haxton

Keywords:

Alpha decay

Geiger–Nuttall law

Formation probability

Clustering

ABSTRACT

The Geiger–Nuttall (GN) law relates the partial α -decay half-life with the energy of the escaping α particle and contains for every isotopic chain two experimentally determined coefficients. The expression is supported by several phenomenological approaches, however its coefficients lack a fully microscopic basis. In this paper we will show that: (1) the empirical coefficients that appear in the GN law have a deep physical meaning, and (2) the GN law is successful within the restricted experimental data sets available so far, but is not valid in general. We will show that, when the dependence of logarithm values of the α formation probability on the neutron number is not linear or constant, the GN law is broken. For the α decay of neutron-deficient nucleus ^{186}Po , the difference between the experimental half-life and that predicted by the GN law is as large as one order of magnitude.

© 2014 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/3.0/>). Funded by SCOAP³.

One landmark in modern physics, shaping the developments leading to Quantum Mechanics, was the formulation of the empirical Geiger–Nuttall (GN) law in 1911 [1]. According to the GN law as formulated in Ref. [1], the α decay partial half-life $T_{1/2}$ is given by

$$\log_{10} T_{1/2} = A(Z)Q_{\alpha}^{-1/2} + B(Z), \quad (1)$$

where Q_{α} is the total energy of the α decay process (α -decay Q value) and $A(Z)$ and $B(Z)$ are the coefficients which are determined by fitting experimental data for each isotopic chain. The GN law has been verified in long isotopic chains and no strong deviations have been observed: It is extremely successful and is considered to be generally valid. Recently the amount of α decay data in heavy and superheavy nuclei has greatly increased [2–7] and the GN law is still fulfilled, reproducing most experimental data within a factor 2–3, as seen in Fig. 1(a) for the Yb–Ra region (apart from the Po chain, as will be discussed in this paper). The coefficients A and B give rise to different GN lines for each isotope series (Fig. 1(a)). The coefficients change for each isotopic chain which crosses the magic numbers, e.g. $N = 126$ [8].

The greatest challenge was thus to understand how the α particle could leave the mother nucleus without any external agent

disturbing it. The first successful theoretical explanation was given by Gamow [9] and independently by Condon and Gurney [10], who explained α decay as the penetration (tunneling) through the Coulomb barrier, leading to the $Q_{\alpha}^{-1/2}$ dependence of Eq. (1). This was a great revolution in physics and confirmed the probabilistic interpretation of Quantum Mechanics. Besides its pioneering role in the development of quantum theory, the α decay also broadens our understanding of the quantum tunneling process of other composite objects. This is a general physical process that can be found in other fields including condensed matter, molecular physics and astrophysics [11–13].

The remaining challenge is to identify the microscopic basis of the GN coefficients A and B . Thomas provided the first attempt of a microscopic theory [14]. His expression for the half life considered the probability that the four nucleons, which eventually constitute the α particle, get clustered at a certain distance on the nuclear surface. In this paper we will apply Thomas's expression to probe the general validity of the GN law, explain the Z dependence of the constants and their microscopic origin.

Guided by recent experimental findings, we divide the α -decaying nuclei into four regions (see Fig. 2):

- (I) $N \leq 126$, $Z \leq 82$;
- (II) $N \leq 126$, $Z > 82$;
- (III) $N > 126$, $Z > 82$;
- (IV) $N > 126$, $Z \leq 82$.

* Corresponding author.

E-mail address: chongq@kth.se (C. Qi).

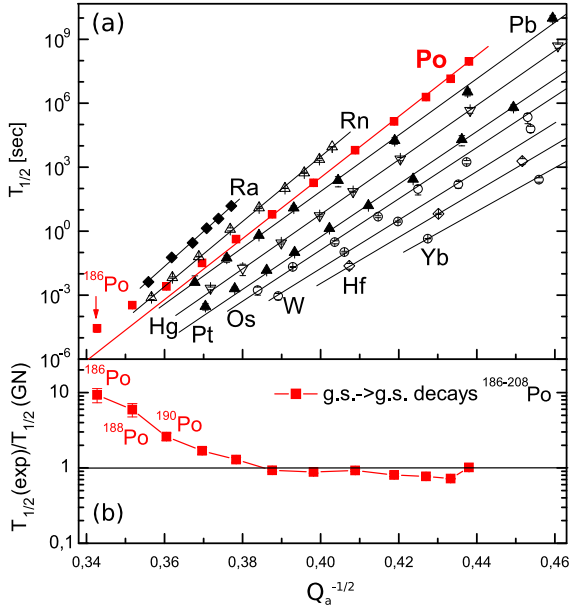


Fig. 1. (a) The logarithms of experimental partial α -decay half-lives (in s) [2,3,7,6] for the even-even Yb–Ra nuclei with neutron number $N < 126$ as a function of $Q_{\alpha}^{-1/2}$ (in $\text{MeV}^{-1/2}$). The straight lines show the description of the GN law with A and B values fitted for each isotopic chain. (b) The deviation of the experimental α -decay half-lives from those predicted by the GN law for the light Po isotopes.

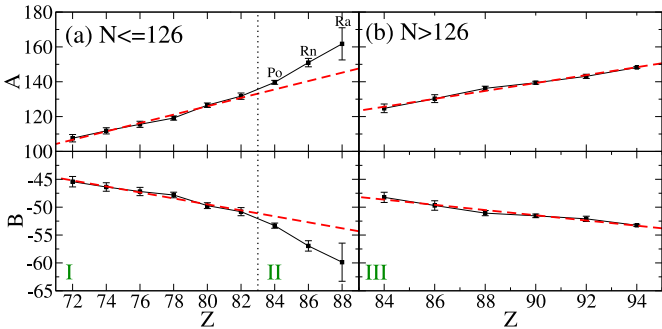


Fig. 2. (Color online.) (a) The coefficients $A(Z)$ and $B(Z)$ for even-even nuclei in regions I ($Z \leq 82$) and II ($Z > 82$) with $N \leq 126$. The red dashed lines are fitted only for the data from region I with $Z \leq 82$ giving $A(Z) = 2.41Z - 66.7$ and $B(Z) = -0.54Z - 6.61$. (b) Same as (a) but for nuclei in region III ($Z > 82$), i.e., polonium to plutonium isotopes with neutron numbers $N > 126$. Again, the red dashed lines are determined by a fitting procedure, which gives $A(Z) = 2.27Z - 65.0$ and $B(Z) = -0.47Z - 9.36$.

Except for ^{210}Pb , α decay has not yet been observed for nuclei in region IV. Fig. 2 shows that both coefficients A and B are linearly dependent upon Z for regions I and III, however with different coefficients. This was initially reported in Ref. [15] and attributed to the crossing of the $N = 126$ neutron shell. The recent extension of the available data also shows that, when crossing the $Z = 82$ shell gap (from region I to region II), another set of coefficients, strongly deviating from the values in regions I and III, is needed. Further, as seen in Fig. 1(b), recent α -decay experiments at SHIP in GSI (Darmstadt, Germany) to study the neutron-deficient Po isotopes [7] show a significant and gradually increasing deviation from the GN law using the coefficients as reported in Fig. 2. In ^{186}Po , the difference between the experimental half-life and that predicted by the GN law is as large as one order of magnitude. Such a strong deviation has not been seen before.

In order to understand the three issues mentioned above we go through the derivations of Refs. [16,17] where a generalization of the GN law was found. This generalization holds for all isotopic

chains and all cluster radioactivities [18]. According to Ref. [14], the α -decay half-life can be written as

$$T_{1/2} = \frac{\ln 2}{\nu} \left| \frac{H_l^+(\chi, \rho)}{R F_{\alpha}(R)} \right|^2, \quad (2)$$

where ν is the velocity of the emitted α particle which carries an angular momentum l . As only ground-state to ground-state α decays of even-even nuclei are considered here, l is equal to 0 in all cases. R is a distance around the nuclear surface where the wave function describing the cluster in the mother nucleus is matched with the outgoing cluster + daughter wave function. H_l^+ is the Coulomb–Hankel function with $\rho = \mu \nu R / \hbar$ and $\chi = 4Ze^2 / \hbar \nu$, the Coulomb parameter, where μ is the reduced mass and Z is the charge number of the daughter nucleus. The quantity $F_{\alpha}(R)$ is the formation amplitude of the α cluster at distance R . Introducing the quantities $\chi' = 2Z\sqrt{A_{\alpha d}/Q_{\alpha}}$ and $\rho' = \sqrt{2A_{\alpha d}Z(A_d^{1/3} + 4^{1/3})}$ where $A_{\alpha d} = 4A_d/(4 + A_d)$, one gets, after imposing the condition of the half life being independent on R [16]

$$\begin{aligned} \log T_{1/2} &= a\chi' + b\rho' + c \\ &= 2aZ/\sqrt{A_{\alpha d}Q_{\alpha}}^{-1/2} + b\sqrt{2A_{\alpha d}Z(A_d^{1/3} + 4^{1/3})} + c, \end{aligned} \quad (3)$$

where a , b and c are constant parameters which only depend upon local variations of the formation probability. They are determined by fitting experimental data [16].

The reason why these parameters are practically constant is that, when going from one isotope to another, the α -particle formation probability usually varies much less than the penetrability. On the logarithm scale of the GN law the differences in the formation probabilities are usually small fluctuations along the straight lines predicted by that law. In other words, the constancy of the parameters a , b and c is a consequence of the smooth variation in the nuclear structure that is often found when going from a nucleus to its neighbors. This is also the reason why, for example, the BCS approximation works so well in many regions of nuclei.

The term $a\chi'$ takes into account the tunneling through the Coulomb barrier, while $b\rho' + c$, which does not depend upon Q_{α} , includes effects induced by the clusterization in the mother nucleus [16]. By comparing equations (1) and (3), a correspondence between the coefficients $A(Z)$ and $B(Z)$ and the expressions $a\chi'$ and $b\rho' + c$ respectively can be deduced and the meaning of the coefficients can be unfolded. The observed linear dependency of $A(Z)$ upon Z is substantiated by this representation. The observed negative values for $B(Z)$ are understood as both terms b and c are negative [16]. The linear dependence upon Z of $B(Z)$ seems to be in conflict with the $Z^{1/2}$ dependence of the term ρ' . However for nuclei with known α -decay half-lives for which the GN law has so far been applied, ρ' is practically a linear function of Z , as seen in Fig. 3.

The need for a different linear Z dependence of the coefficients A and B in the four regions of the nuclear chart (see Fig. 2) will now be addressed. A generic form for the evolution of the alpha formation probabilities was proposed in [7]. It was based on experimental values [3,4,6,7] and calculations performed within the framework of the seniority scheme. This generic form is shown in Fig. 4 for selected isotopic chains. Three distinct features can be extracted from this schematic representation.

The experimental α formation probabilities of most known α emitters in regions I and III are nearly constant as a function of neutron number (or more exactly, weakly linearly dependent on ρ' , as seen in Fig. 1 of Ref. [19]). For those nuclei, the GN law is indeed expected to be valid and $A(Z)$ and $B(Z)$ follow a linear behavior as a function of Z (see Figs. 1 and 3 and Eq. (9) in Ref. [17]).

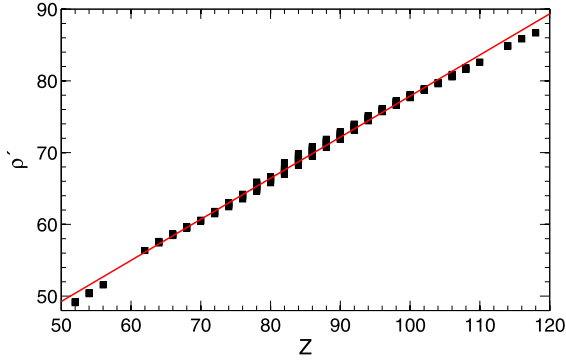


Fig. 3. (Color online.) Calculated values of ρ' for observed even-even α emitters as a function of Z . The linear fit to the values is shown by the solid line.

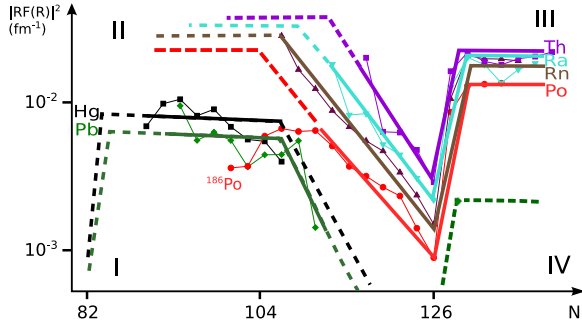


Fig. 4. A pictorial representation of the generic form of the evolution of the α formation probabilities $|RF(R)|^2$. Thick solid lines are for isotopes, where experimental data are available and dashed lines are extrapolations to the regions with the yet unavailable data. The experimental data as cited in [7] are shown by points, connected by thin lines, to guide the eye.

Approaching the $N = 126$ shell closure with increasing neutron number, a strong, exponential decrease of the formation probability is observed (see Fig. 4, in region II and the discussion in Ref. [7]). It is striking that in spite of a variation of $|RF_\alpha(R)|^2$ over one order of magnitude, the GN law and the $A(Z)$ and $B(Z)$ linear dependence upon Z are still valid. This has no real physical meaning, but is a consequence of the specific dependence of the $|RF_\alpha(R)|^2$ on Q_α . The Q_α (as well as $Q_\alpha^{-1/2}$) values exhibit a quasi-linear pattern as a function of rising neutron number when approaching the $N = 126$ shell closure. Therefore $\log_{10}|RF(R)|^2$ and thus $\log_{10}(T_{1/2})$ will still depend linearly on $Q_\alpha^{-1/2}$. As examples, in Fig. 5 the logs of the α formation probabilities $|RF(R)|^2$ for polonium and radon isotopes in regions II and III are shown as a function of $Q_\alpha^{-1/2}$. In comparison with those in region III for which the formation probabilities are nearly constant or only weakly depend on $Q_\alpha^{-1/2}$, the data in region II show an exponential dependence. The other isotopic chains in region II show a similar linearly decreasing behavior of $\log_{10}|RF(R)|^2$ as a function of $Q_\alpha^{-1/2}$, as indicated by the red-dashed lines in the figure, however with different slopes. As a result, the GN law remains valid for isotopic chains in region II, but the corresponding values of A and $|B|$ will increase with Z beyond the trend observed in regions I and III (see Fig. 2).

For the polonium isotopic chain with $N < 126$, the linear behavior of $\log_{10}|RF(R)|^2$ breaks down below ^{196}Po ($N = 112$, corresponding to $Q_\alpha^{-1/2} = 0.39$ in Fig. 5). This explains why the GN law is broken in the light polonium isotopes of Fig. 1(b). This violation of the GN law, observed here for the first time, is induced by the strong suppression of the α formation probability due to the fact that the deformations and configurations of the ground

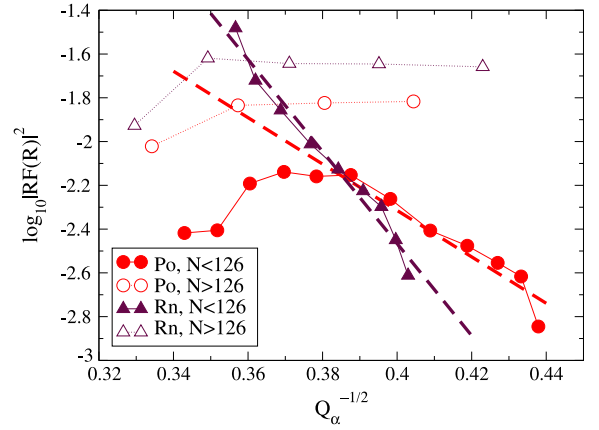


Fig. 5. (Color online.) $\log_{10}|RF(R)|^2$ for Po (circle) and Rn (triangle) isotopes in region II with $N < 126$ (closed symbols) and region III with $N > 126$ (open symbols) as a function of $Q_\alpha^{-1/2}$. The dashed lines are to guide the eye.

states of the lightest α -decaying neutron-deficient polonium isotopes ($A < 196$) are very different from those of the daughter lead isotopes [20,21]. It should be mentioned that our generic form on the evolution of $F(R)$ presented here and in Ref. [7] is mainly guided by available experimental data. A systematic microscopic calculation on $F(R)$ is desired to confirm this conjecture.

In conclusion, we have studied the origin and physical meaning of the coefficients $A(Z)$ and $B(Z)$ in the GN law. These coefficients are determined from experimental data and show a linear dependence upon Z . However, the Z -dependence is different in different regions of the nuclear chart. Starting from the microscopic Thomas expression for the decay half life we show that $A(Z)$ models the tunneling process as well as the relatively small variations in the structure of the neighboring nuclei. The parameter $B(Z)$ takes into account the clusterization of the α -particle in the mother nucleus. We show why the coefficient $B(Z)$ is negative and that both $A(Z)$ and $B(Z)$ have to be practically linearly dependent upon Z . We also demonstrated here for the first time that, when the dependence of $\log_{10}|RF(R)|^2$ on the neutron number is not linear or constant, the GN law is broken. This also explains why the GN law works so well in all α emitters known today except for the polonium isotopes, as the data within each isotopic chain are so far limited to a region where $\log_{10}|RF(R)|^2$ behaves linearly with N or is constant. It is only for the polonium isotopic chain that experimental data have been obtained over a wide enough range to observe significant deviations from the GN law. Within the generic description [7], the different values of the alpha formation probability for regions I and III and the exponential decrease as a function of neutron number when approaching $N = 126$ for region II, can be understood as due to the available j orbitals and a difference in the clustering properties of the nucleons in the α particle. Clustering of the two protons and two neutrons leading to the α -particle formation proceeds through high-lying empty single particle configurations. It would therefore be very interesting to extend the experimental knowledge towards more neutron deficient radon, radium and thorium isotopes in region II and to more neutron-rich lead and mercury isotopes in regions I and IV. This will allow us to validate the generic description, identify the saturation levels of the α formation probability and to investigate the influence of protons and neutrons filling the same single particle orbitals (between 82 and 126) [22–24]. Consequently, compared to the use of the so far generally accepted GN description, more reliable predictions of the α decay half lives will be achieved in unknown nuclei and in low α -decay branching ratios close to stability.

Acknowledgements

This work has been supported by the Swedish Research Council (VR) under grant Nos. 621-2012-3805 and 621-2013-4323, FWO Vlaanderen (Belgium), GOA/2004/03 (BOF K.U. Leuven), the IUAP – Belgian State – Belgian Science Policy (BriX network P7/12), and by the European Commission within the Sixth Framework Programme through I3-ENSAR (contract No. RII3-CT-2010-262010) and by the STFC of UK.

References

- [1] H. Geiger, J.M. Nuttall, *Philos. Mag.* 22 (1911) 613;
H. Geiger, *Z. Phys.* 8 (1922) 45.
- [2] G. Audi, O. Bersillon, J. Blachot, A.H. Wapstra, *Nucl. Phys. A* 729 (2003) 3;
G. Audi, et al., *Chin. Phys. C* 36 (2012) 1157.
- [3] NNDC at the Brookhaven National Laboratory, <http://www.nndc.bnl.gov/nudt2/>.
- [4] A.N. Andreyev, et al., *J. Phys. G, Nucl. Part. Phys.* 37 (2010) 035102.
- [5] M. Pfützner, M. Karny, L.V. Grigorenko, K. Riisager, *Rev. Mod. Phys.* 84 (2012) 567.
- [6] K. Van de Vel, et al., *Phys. Rev. C* 68 (2003) 054311.
- [7] A.N. Andreyev, et al., *Phys. Rev. Lett.* 110 (2013) 242502.
- [8] B. Buck, A.C. Merchant, S.M. Perez, *Phys. Rev. Lett.* 65 (1990) 2975.
- [9] G. Gamow, *Z. Phys.* 52 (1929) 510.
- [10] E.U. Condon, R.W. Gurney, *Nature (London)* 122 (1928) 438;
R.W. Gurney, E.U. Condon, *Phys. Rev.* 33 (1929) 127.
- [11] A.B. Balantekin, N. Takigawa, *Rev. Mod. Phys.* 70 (1998) 77.
- [12] M. Razavy, *Quantum Theory of Tunneling*, World Scientific, 2003.
- [13] A. Lemasson, et al., *Phys. Rev. Lett.* 103 (2009) 232701.
- [14] R.G. Thomas, *Prog. Theor. Phys.* 12 (1954) 253.
- [15] V.E. Viola, G.T. Seaborg, *J. Inorg. Nucl. Chem.* 28 (1966) 741.
- [16] C. Qi, F.R. Xu, R.J. Liotta, R. Wyss, *Phys. Rev. Lett.* 103 (2009) 072501.
- [17] C. Qi, F.R. Xu, R.J. Liotta, R. Wyss, M.Y. Zhang, C. Asawatangtrakuldee, D. Hu, *Phys. Rev. C* 80 (2009) 044326.
- [18] C. Qi, D.S. Delion, R.J. Liotta, R. Wyss, *Phys. Rev. C* 85 (2012) 011303(R).
- [19] C. Qi, A.N. Andreyev, M. Huyse, R.J. Liotta, P. Van Duppen, R.A. Wyss, *Phys. Rev. C* 81 (2010) 064319.
- [20] A.N. Andreyev, M. Huyse, P. Van Duppen, J.F.C. Cocks, K. Helariutta, H. Kettunen, P. Kuusiniemi, M. Leino, W.H. Trzaska, K. Eskola, R. Wyss, *Phys. Rev. Lett.* 82 (1999) 1819.
- [21] D. Karlgren, R.J. Liotta, R. Wyss, M. Huyse, K. Van de Vel, P. Van Duppen, *Phys. Rev. C* 73 (2006) 064304.
- [22] S.N. Liddick, et al., *Phys. Rev. Lett.* 97 (2006) 082501.
- [23] S.N. Liddick, et al., *Eur. Phys. J. Spec. Top.* 150 (2007) 131.
- [24] D. Seweryniak, et al., *Phys. Rev. C* 73 (2006) 061301.